

Evolution of temporal coherence in confined polariton condensates

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Abstract: We study the second order temporal coherence of the emission from a semiconductor microcavity in the strong coupling regime. We evidence the favorable influence of spatial confinement, realized by etching micropillar structures, on the temporal coherence of solid state quasi-condensates which evolve in our device above threshold. By fitting the experimental data with a microscopic quantum theory based on a Monte Carlo wavefunction approach, we scrutinize the influence of the crystal lattice temperature (interaction with acoustic phonons) and pump power on the condensate's temporal coherence. Phonon-mediated transitions in the optical mode structure are observed, which in the case of a confined structure splits into a set of discrete resonances. By increasing the pump power beyond the condensation threshold, the temporal coherence significantly improves in the pillar devices, as revealed in the transition from thermal to coherent statistics of the emitted light.

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OCIS codes: (250.5590) Quantum-well, -wire and -dot devices; (190.5890) Scattering, stimulated; (230.5750) Resonators; (140.3948) Microcavity devices

1. Introduction

The temporal coherence of a source of radiation is a key quantity which discriminates a laser-like device from thermal emitters. While the first order coherence function, $g^{(1)}(\tau)$, which is the correlator of field amplitudes at different times, reflects the coherence of the emitted photons, its second order counterpart $g^{(2)}(\tau)$, that involves intensity correlation, gives insights into the emission statistics. In conventional semiconductor lasers which rely on stimulated emission and population inversion, the emission statistics transits from thermal below lasing threshold to coherent as revealed by $g^{(2)}(\tau = 0) = 2$ and 1 respectively. Even in the limiting case of a vanishing threshold occurring in devices where spontaneous emission is perfectly funneled into the lasing mode, laser oscillation can still be identified since $g^{(2)}(0)$ approaches a value of 1 with increasing pump power [1].

An example of a light emitting device which may operate as a coherent light source without population inversion, is a semiconductor microcavity in the strong light–matter coupling regime [2]. The strong coupling results in the emergence of hybrid eigen modes called exciton-polaritons (later, *polaritons*) [3, 4]. Being very low-mass bosons, they can undergo a dynamic Bose-Einstein condensation (BEC) process at elevated temperatures. Analogous to a conventional laser operating in the weak coupling regime (for instance the vertical cavity surface emitting laser, VCSEL), the formation of a BEC of polaritons is accompanied by a nonlinear increase of the intensity of the emitted light and a drop of the spectral linewidth [2]. The latter is a typical, yet not unambiguous signature of first order temporal coherence of the emitted radiation. A more sophisticated approach relying on Michelson interferometry was discussed in [5], where strongly enhanced coherence times in the regime of polariton lasing have been demonstrated by using low noise pump sources. In analogy to cold atom BECs, the first order spatial coherence (long range order), has been considered as a smoking gun criterion for the claim of a polariton BEC [6–9]. However, the second order coherence function which represents another key signature of coherent light, is less well understood. One difficulty arises from the lack of accurate theoretical quantum description of the behavior of $g^{(2)}(\tau)$ in real conditions, such as finite temperatures. Indeed, common methods are based on stochastic Gross-Pitaevskii [10–12] equations or expectation values evolution [13] that rely on severe assumptions regarding high order correlations such as coherence functions. Additionally, the $g^{(2)}(\tau \neq 0)$ dependence could not be addressed by semiclassical models. As a consequence of these complications, reports on the second order coherence function in polariton systems have not been fully conclusive in unified experimental theoretical works. A second difficulty is based on the results of present experimental works: Here, in most cases, a surprisingly slow drop down of $g^{(2)}(0)$ above threshold

is commonly observed in two-dimensional microcavities [14–16] with a persisting value significantly above unity, thus implying a strong deviation from a Poissonian photon statistics. Even an increase of $g^{(2)}(0)$ with increasing pump power has been reported, both for GaAs and CdTe based samples [15, 17, 18]. It is reasonable to assume that these peculiarities are a result of a large number of states in the continuous dispersion of polaritons confined in planar microcavities, which can contribute to the condensation phenomena and unavoidably lead to mode competition effects. Such effects further convolute the coherence phenomena inherent to the polariton condensation, and therefore are considered as another main obstacle towards a precise understanding of the evolution of second-order temporal coherence in solid state condensates beyond a phenomenological level.

In this article we present a systematic combined experimental and theoretical study of the power and temperature dependence of the $g^{(2)}$ function both in confined and planar strongly coupled microcavity structures. Spatial confinement has been used to enhance condensation processes in trap structures and micropillars[19–21] with a good overview given in [22]. In our case spatial confinement in etched micropillars allows us to form a single mode polariton condensate, thus $g^{(2)}(0)$ converges towards unity for large pump powers. Our results are supported by a stochastic quantum trajectory approach which provides unprecedented insight into the interplay between state occupation, particle fluctuations, and photon coherence. In particular, we show that accounting for dissipative polariton-phonon interaction and polariton self interactions in a fully quantum manner is sufficient to retrieve the (resolution limited) $g^{(2)}(0)$ behavior in confined structures where a discrete dispersion assists the condensation.

2. Experimental details

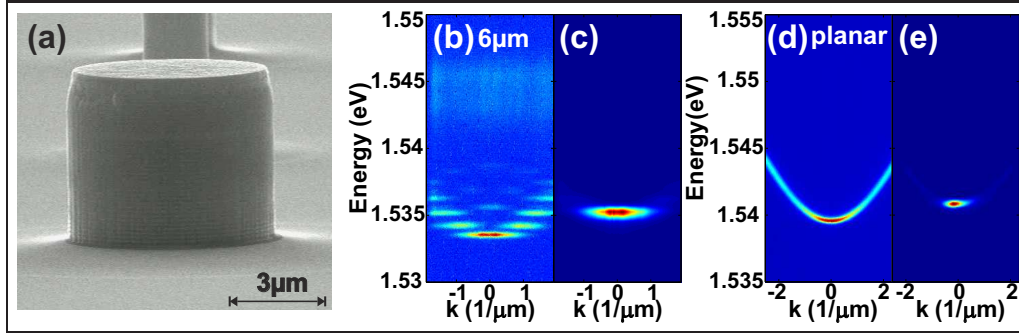


Fig. 1. (a) Scanning electron microscope picture of the processed micropillars. Energy dispersion measured in PL for the pillar (b) far below and (c) above threshold. Emission from the planar sample (d) far below and (e) above threshold.

The sample which we study is a high-Q AlGaAs alloy-based $\lambda/2$ planar microcavity with twelve GaAs quantum wells (QWs) of 13 nm width each located in the optical antinodes of the confined electromagnetic field. The design of the sample is similar to the one described in [23]. In order to enhance the Q-factor, we increased the number of AlGaAs/AlAs mirrorpairs in the top(bottom) distributed Bragg reflector (DBR) segment to 23(27). The Q-factor was experimentally estimated to exceed 10000 for highly photonic structures.

To provide lateral confinement of the polariton modes, we fabricated micropillars with the diameters 6 μm . Electron beam lithography and electron-cyclotron-resonance reactive-ion-etching were used for deeply etching the cavity. A scanning electron microscope (SEM) image of the sample is shown in Fig. 1(a). We also extracted the Rabi-splitting of the device via white

light reflection and found a value of 10.1 meV for the heavy hole-based exciton [16].

The sample is pumped with a pulsed Ti:Sa laser tuned to the first Bragg minimum of the stop band located approximately 100 meV above the free exciton resonance. The pulse width was 50 ps.

It is worth noting, that in most of the previous studies devoted to the second-order correlation function in polariton systems, very short pumping pulses of 1-3 ps have been used.

On one hand, short pulsing is advantageous over long-pulse excitation schemes since conventional single photon detectors with time resolution of the order of hundreds ps can be used to extract the second order correlation function at zero time delay $g^{(2)}(0)$ [17, 23]. The finite length of the emission pulse effectively acts as a time filter in this configuration [23] as only photons which are emitted within the lifetime of the condensate and reservoir are correlated.

On the other hand, it has been realized by now that fundamental behavior of a polariton laser (in terms of its linewidth and emission frequency) is drastically modified by the ultra short excitation. It results in significant line broadenings and energy shifts [24], which strongly affects the coherence properties. As our pulse width is far below the resolution of our detectors of 400 ps and still much shorter than the pulse repetition period of around 12 ns, our excitation scheme can be considered as a compromise between reasonable time resolution and keeping equilibrium conditions in the experiment. For excitation beam diameters smaller than the pillar width, we gain high sensitivity of the output to the spatial position of the laser leading to excitement of desired optical modes [25]. However, using narrow beams breaks the homogeneity of the pumping scheme. Therefore small pumping spots lead to a repulsive potential driving the polaritons away from the center, i.e. out of the ground state, causing condensation at high energies and k vectors [26]. To suppress this effect and ensure a homogeneous excitation of the sample, we expanded the beam diameter to $40\mu\text{m}$. This value is more than six times bigger than the pillar width. Moreover, to further improve the homogeneity of the excitation, a closed optical aperture and a lens were used for beamshaping. The dispersions were recorded using a 4f Fourier setup [27].

3. Experimental results and discussion

Tight optical confinement provided by the micropillar landscape gives rise to a characteristic set of discrete optical modes in the lower polariton dispersion, see Fig. 1(b). On top of the mode spectrum, we detect an emission band around 1.545 eV which we attribute to radiation of free excitons which scatters through the sidewalls of the pillars. In contrast, for the case of the planar (not etched) structure, the dispersion has a continuous parabolic shape, see Fig. 1(d), and the free exciton radiation is completely suppressed. We also note, that the energy of free excitons is somewhat blue shifted compared to the planar sample as a result of the modified strained environment. Above a distinct threshold occurring at pump powers around 45 W/cm^2 , both for the planar and the structured sample, we observe a massive occupation of the polariton ground state at the bottom of the lower polariton branch, indicating the formation of a polariton condensate. The spectra in Fig. 1(c,e) were recorded at injection powers of ca. 50 W/cm^2 . Additionally the dispersion for 5 (lower row) and 40 K (upper row) for different excitation powers are shown in Fig. 2. The transition from the mode structure emission for low powers ((a) and (d)) to the slight broadening of the fundamental mode shortly before threshold ((b) and (e)) and the ground state condensation above threshold ((c) and (f)) can be seen for both temperatures.

In order to give unambiguous evidence for polariton lasing in our quasi-zero-dimensional system, we carefully investigate the luminescence from the ground state of a pillar with a detuning $\Delta(40\text{K}) = -8 \pm 0.5\text{ meV}$ at a sample temperature of 40 K. The dispersion is integrated from $k = -0.3\mu\text{m}$ to $k = 0.3\mu\text{m}$ and fitted by a Lorentzian. We observe a distinct thresh-

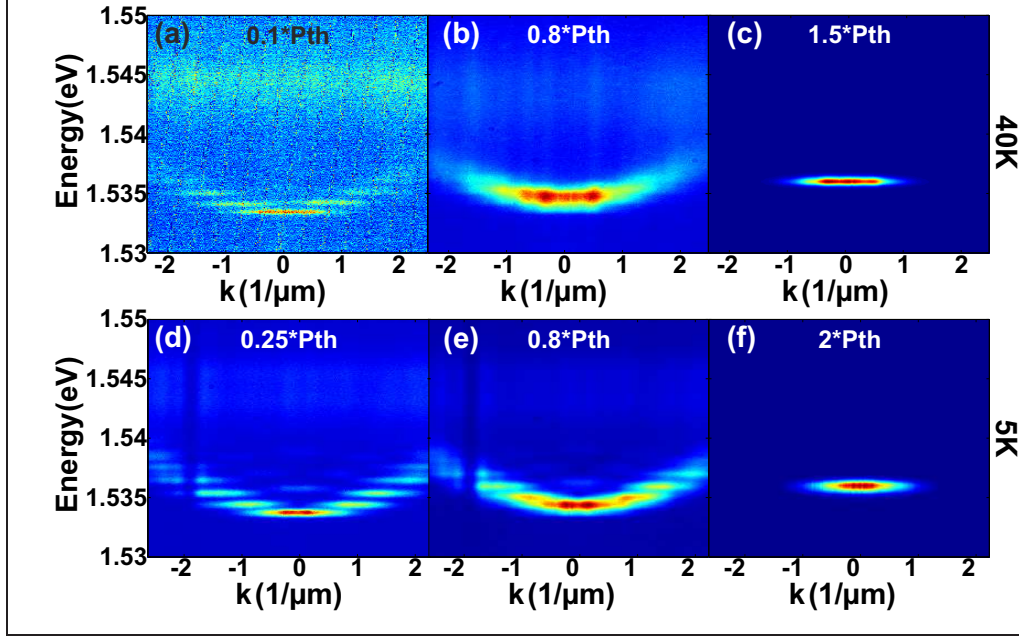


Fig. 2. False color plots of the energy dispersion for 40K(upper row) and 5K (lower row) (a) and (d) far below (b) and (e) slightly below and (c) and (f) above threshold. (b) and (e) show a somewhat broadened energy ground state, resulting from the pulsed excitation conditions.

old behaviour in the input-output-characteristic with a non-linearity occurring at 45 W/cm^2 , see Fig. 3(a). While the energy of the ground state features a linear blueshift with increasing pump power below threshold (caused by interaction with the exciton reservoir), the blueshift becomes logarithmic as soon as the threshold is reached. This behaviour displayed in Fig. 3(c) is in agreement with data reported in [28] and indicates that repulsive polariton-polariton interactions play a dominant role. Furthermore, we clearly observe narrowing of the emission linewidth (FWHM) at the threshold, also shown in Fig. 3(c) indicating the transition from thermal to coherent emission of light. The behaviour for the pillar at 5 K is similar and therefore not shown. Fig. 3(b) and (d) show analogous data for the planar measurement. The distinct threshold in Fig. 3(b), accompanied by the continuous blueshift above and the drop of the linewidth at the threshold in Fig. 3(d) prove polaritonic condensation also in the planar regime.

Further, we study the temporal second-order correlation function,

$$g^{(2)}(\tau) = \frac{\langle I(t+\tau)I(t) \rangle}{\langle I(t) \rangle \langle I(t+\tau) \rangle}, \quad (1)$$

where $I(t)$ is the emission intensity at time t and $\langle \dots \rangle$ means time averaging. For the photon statistics measurements, we use a Hanbury-Brown and Twiss configuration with two avalanche photodiodes having a time resolution of 400 ps each [16, 17, 23]. The light emitted from the sample is filtered by a monochromator with a resolution of 0.2 meV to avoid errors caused by interaction with higher-energy states.

Figure 4(a) shows a zoom onto the $\tau = 0$ peak for the planar sample (black) and the pillar sample (red) far above ($10.5 * P_{th}$) threshold. The difference in height of the $\tau = 0$ peak visualizes the difference in the $g^{(2)}$ value. The inset shows the corresponding normalized $g^{(2)}$

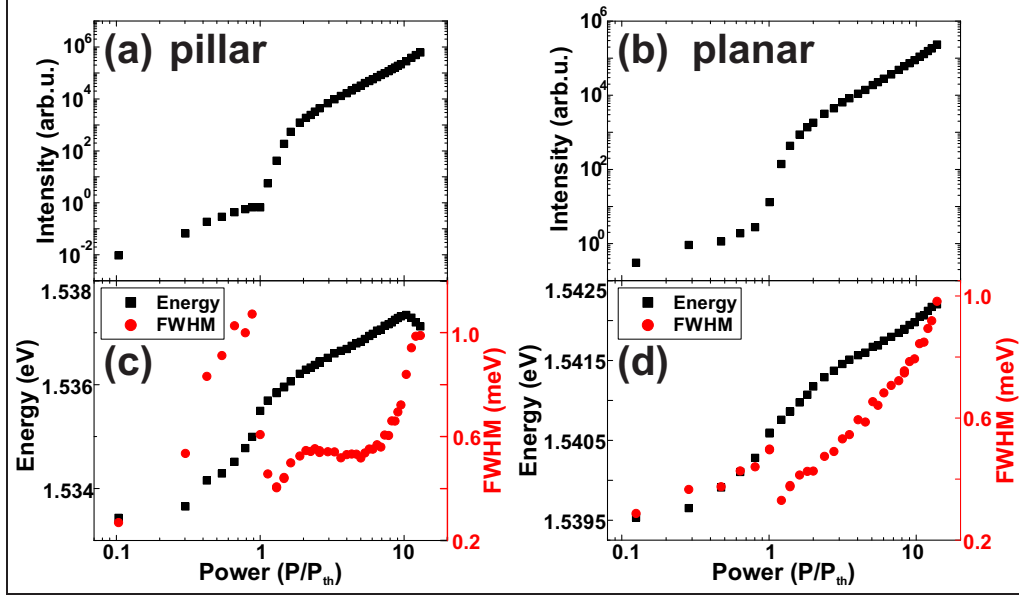


Fig. 3. Ground state input output characteristic with a pronounced non linearity at the threshold for (a) the pillar and (b) the planar structure. Emission energy (black, left axis) and linewidth (red, right axis) vs. pumping power normalized to the threshold for (c) the pillar and (d) the planar structure. The persisting blueshift above threshold and the linewidth drop at the threshold evidence the formation of a polariton condensate.

measurements. The data has been normalized to the average of the side peaks. Figure 4(b) analogously shows the same behaviour for 5K (red) and 40K (black) measured on the pillar sample. The inset clearly shows a $g^{(2)}$ value bigger than 1 for both measurements.

Figure 4(c) shows the power dependence of $g^{(2)}(0)$ in the planar microcavity and the pillar structure for a detuning of $\Delta(5K) = -8.5 \pm 0.5 \text{ meV}$. Below threshold one would expect $g^{(2)}(0) = 2$ for thermal emission. Since the temporal resolution of the measurements exceeds the temporal correlation time of the emitter this can not be resolved leading to $g^{(2)}(0) = 1$ [29, 30]. The increase of the central peak around $\tau = 0$ at the threshold can be attributed to an enhanced two-photon emission probability and indicates that a fully coherent state is not yet formed. Far above threshold the system has reached a coherent state represented by $g^{(2)}(0) = 1$. To estimate the actual value of $g^{(2)}(0) = N_0/\bar{N}_S$, the integrated intensity of the $\tau = 0$ peak, N_0 , is divided by the average intensity of the $\tau \neq 0$ peak, $\bar{N}_S = N_S/n$, where n is the number of side peaks.

In the planar case, $g^{(2)}(0)$ is significantly increased at large excitation powers, and remains almost constant at a value around $g^{(2)}(0) \approx 1.1$ for increasing pump power. This is in qualitative agreement with previous reports [15–17, 23]. The effect has been assigned to a reservoir depletion triggered by polariton self-scattering from the ground state into the higher-energy states. An alternative interpretation of such behavior reverts to multimode polariton lasing from the ground state and existence of a finite set of emitting k -values, which leads to an increase of $g^{(2)}(0)$.

In the structure with pillars, both of the above mentioned effects should be strongly suppressed due to the discrete mode spectrum and optical confinement. As shown in Fig. 3, the pillar device exhibits single-mode lasing on the fundamental mode. The mode spacing from the ground state to the next higher state amounts to around 0.8 meV. This should drastically

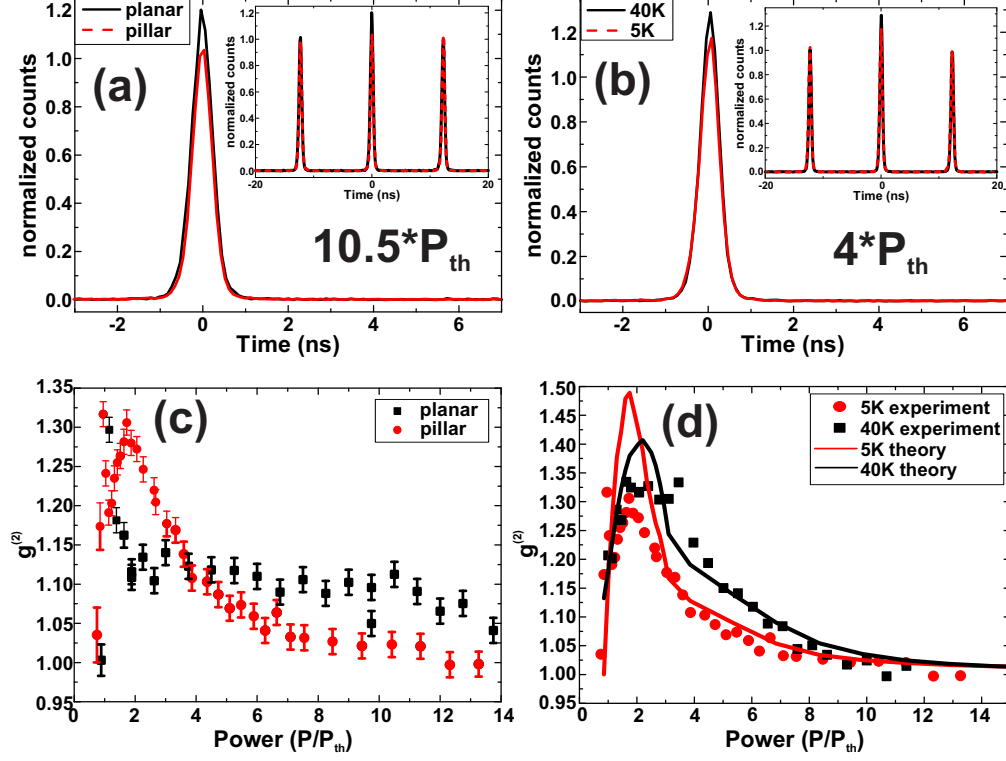


Fig. 4. (a) Zoom onto the $\tau = 0$ peak for the planar sample (black) and pillar sample (red) taken at $10.5 * P_{th}$. Inset shows corresponding normalized $g^{(2)}$ measurements (b) Zoom onto the $\tau = 0$ peak for 40K (black) and 5K (red) taken at $4 * P_{th}$ on the pillar sample. Inset shows corresponding normalized $g^{(2)}$ measurements (a) and (b) visualize the difference in the $g^{(2)}$ value. (c) Comparison between pillar (red circles) and planar (black squares) emission: For large pump powers, $g^{(2)}(0)$ drops significantly faster in the micropillar as a result of spatial confinement. (d) $g^{(2)}(0)$ dependence on the pumping power for the pillar sample at 5K (red circles) and 40K (black squares) with fit (line). The enhanced phonon effects for higher temperatures cause a slower drop of the $g^{(2)}(0)$ -value.

reduce the condensate depletion effect at cryogenic temperatures [31]. We directly evidence this improvement by observing a significantly faster drop of the $g^{(2)}(0)$ towards unity in the micropillar structure, see Fig. 4(c).

We have investigated the power dependent behaviour of $g^{(2)}(0)$ for temperatures of 5 and 40 K in Fig. 4(d) in the same $6\mu\text{m}$ pillar device. With increasing temperature, the drop of $g^{(2)}(0)$ towards 1 successively becomes slower, which suggests a phonon induced condensate depletion effect, leading to enhanced number of fluctuations even in the presence of optical confinement.

4. Theoretical description

The system is described by the Hamiltonian

$$\mathcal{H} = \mathcal{H}_{\text{kin}} + \mathcal{H}_{\text{p-p}} + \mathcal{H}_{\text{p-ph}} + \mathcal{H}_{\text{pump}}. \quad (2)$$

The first two terms: free propagation term, \mathcal{H}_{kin} , and polariton-polariton elastic scattering term, $\mathcal{H}_{\text{p-p}}$, encompass the coherent processes. The last two terms namely the coupling to a thermal

bath of acoustic phonons, $\mathcal{H}_{\text{p-ph}}$, and the nonresonant excitation, $\mathcal{H}_{\text{pump}}$, are written using the rotating wave approximation and correspond to incoherent processes. The incoherent processes are treated through a Monte Carlo wave function approach [32, 33] given the corresponding quantum jump operators

$$\hat{\mathcal{J}}_{\mathbf{k}}^+ = \sqrt{\gamma_{\mathbf{k}} \bar{n}_{\text{p}}(E_{\mathbf{k}})} \hat{a}_{\mathbf{k}}^\dagger, \quad (3)$$

$$\hat{\mathcal{J}}_{\mathbf{k}}^- = \sqrt{\gamma_{\mathbf{k}} [\bar{n}_{\text{p}}(E_{\mathbf{k}}) + 1]} \hat{a}_{\mathbf{k}}, \quad (4)$$

$$\hat{\mathcal{J}}_{\mathbf{k}_1 \mathbf{k}_2}^+ = \sqrt{\gamma_{\mathbf{k}_1 \mathbf{k}_2}^{\text{ph}} \bar{n}_{\text{ph}}(E_{\mathbf{k}_1} - E_{\mathbf{k}_2})} \hat{a}_{\mathbf{k}_1}^\dagger \hat{a}_{\mathbf{k}_2}, \quad (5)$$

$$\hat{\mathcal{J}}_{\mathbf{k}_1 \mathbf{k}_2}^- = \sqrt{\gamma_{\mathbf{k}_1 \mathbf{k}_2}^{\text{ph}} [\bar{n}_{\text{ph}}(E_{\mathbf{k}_1} - E_{\mathbf{k}_2}) + 1]} \hat{a}_{\mathbf{k}_1} \hat{a}_{\mathbf{k}_2}^\dagger, \quad (6)$$

where $\hat{a}_{\mathbf{k}}$ are annihilation operators of the polariton modes, $\gamma_{\mathbf{k}}$ is a wave vector-dependent lifetime of polaritons, \bar{n}_{p} is a pumping source distribution, $E_{\mathbf{k}}$ is a polariton dispersion, $E_{\mathbf{k}_1} > E_{\mathbf{k}_2}$, $\gamma_{\mathbf{k}_1 \mathbf{k}_2}^{\text{ph}}$ is a polariton-phonon scattering rate, and \bar{n}_{ph} is a phonon bath distribution.

Using the experimental parameters, we simulate the quantum dynamics of our system initially prepared in a vacuum state. In brief, the procedure consists in evolution of the system wave function through the Schrödinger equation,

$$i\hbar \frac{\partial}{\partial t} |\tilde{\psi}\rangle = \hat{\mathcal{H}}_{\text{eff}} |\tilde{\psi}\rangle, \quad (7)$$

with an effective Hamiltonian

$$\begin{aligned} \hat{\mathcal{H}}_{\text{eff}} = & \hat{\mathcal{H}} - \frac{i\hbar}{2} \sum_{\mathbf{k}} \hat{\mathcal{J}}_{\mathbf{k}}^{+\dagger} \hat{\mathcal{J}}_{\mathbf{k}}^+ - \frac{i\hbar}{2} \sum_{\mathbf{k}} \hat{\mathcal{J}}_{\mathbf{k}}^{-\dagger} \hat{\mathcal{J}}_{\mathbf{k}}^- \\ & - \frac{i\hbar}{2} \sum_{\mathbf{k}_1 \mathbf{k}_2} \hat{\mathcal{J}}_{\mathbf{k}_1 \mathbf{k}_2}^{+\dagger} \hat{\mathcal{J}}_{\mathbf{k}_1 \mathbf{k}_2}^+ - \frac{i\hbar}{2} \sum_{\mathbf{k}_1 \mathbf{k}_2} \hat{\mathcal{J}}_{\mathbf{k}_1 \mathbf{k}_2}^{-\dagger} \hat{\mathcal{J}}_{\mathbf{k}_1 \mathbf{k}_2}^-, \end{aligned} \quad (8)$$

where the non-Hermitian parts results in a decay of the norm $\langle \tilde{\psi}(t) | \tilde{\psi}(t) \rangle$ from unity. Further, a random number, η , is generated. If during the evolution by Eq. (7), the condition $\langle \tilde{\psi}(t) | \tilde{\psi}(t) \rangle \leq \eta$ is met, a jump occurs. After each jump, the state $|\tilde{\psi}(t)\rangle$ should be renormalized and a new number η generated. Each realization, j , of this protocol yields a quantum trajectory, $|\tilde{\psi}(t)\rangle_j$, where $j = 1, 2, \dots, N$. Later, we approximate the density matrix of the system as

$$\hat{\rho}(t) = \frac{\sum_{j=1}^N |\tilde{\psi}(t)\rangle_j \langle \tilde{\psi}(t)|_j}{N} \xrightarrow{N \rightarrow \infty} \hat{\rho}(t), \quad (9)$$

where $\hat{\rho}(t)$ is the density matrix of the system. The approximate expectation value, $\langle \hat{O}(t) \rangle$, of any system observable \hat{O} can be found from

$$\langle \hat{O}(t) \rangle = \text{Tr} [\hat{O} \hat{\rho}(t)] \xrightarrow{N \rightarrow \infty} \text{Tr} [\hat{O} \hat{\rho}(t)] = \langle \hat{O}(t) \rangle. \quad (10)$$

In our computations we account for 2 excited states in addition to the ground state in a truncated Hilbert space [33] of sufficient dimension to avoid particle number saturation for the pump power we consider.

To mimic the finite resolution t_{res} of the HBT setup, we reconstruct the $g^{(2)}(\tau)$ function from the full ground state decay statistics imposed by the action of the operator (3) on the ground state via \hat{a}_0 . Then we reconstruct the resolution limited $g^{(2)}(0)$ as

$$g^{(2)}(0) = \int_0^{t_{\text{res}}} g^{(2)}(\tau) d\tau. \quad (11)$$

As shown in Fig.4(d) that compares theory to experiment for 2 different temperature, we were able to reproduce the drop of $g^{(2)}(0)$ towards 1 quantitatively for the entire investigated temperature range. The discrete dispersion strongly assists the onset of coherence in the system for the reason that a polariton goes down the dispersion (via phonon emission) at a rate proportional to $\bar{n}_{\text{ph}}(E_{\mathbf{k}_1} - E_{\mathbf{k}_2}) + 1 \simeq 1$ while the probability of going up (via phonon absorption) is proportional to $\bar{n}_{\text{ph}}(E_{\mathbf{k}_1} - E_{\mathbf{k}_2}) \ll 1$. Indeed, given the relatively large gaps of about 0.8 meV imposed by the lateral confinement (see Fig.1(b)) the average number of phonons available for such transitions remains much lower than unity and a polariton reaching the ground state can hardly thermalize especially at large pump powers. The saturation of the $g^{(2)}(0)$ to a value of about 1.4 results from the averaging imposed by the HBT resolution. In our simulation before averaging we systematically obtain $g^{(2)}(0) \simeq 2$ or even slightly above at the peak around threshold. Noteworthy, our model is additionally able to reproduce $g^{(2)}(0) \simeq 1$ far below threshold which results from the absence of up stimulation process given the low polariton occupation involved. Finally we underline that the quantitative level discrepancies between theory and experiment could be wiped out adding a new set of quantum jump operators accounting for a weak pure dephasing in the system [34] and therefore slightly reducing the coherence.

These results underline the power of the quantum jump approach to our discrete mode polariton scheme and support our conclusion that the presence of optical confinement strongly improves the coherence of our polariton laser and partly protects the device from phonon induced condensate depletion effects for higher temperatures.

5. Conclusion

We have evaluated the effect of optical confinement and the lattice temperature on the temporal coherence function behavior of a polariton condensate. Experimentally, we observe a significantly faster drop of $g^{(2)}(\tau = 0)$ towards unity above the polariton laser threshold, compared to planar polariton microcavity reference structures. By combining our experimental data with a microscopic model based on the quantum jump approach, we were able to successfully describe the influence of phonons on the coherence evolution in our polariton devices, which is of paramount importance for the design of next generation coherent polariton light sources.

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